

***Interactive comment on* “Consistent simulation of bromine chemistry from the marine boundary layer to the stratosphere, Part I: model description, sea salt aerosols and pH” by A. Kerkweg et al.**

A. Kerkweg et al.

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We thank the anonymous referee #3 for the valuable and constructive comments, to which we reply in the following:

We fully agree that the recycling of bromine on the accumulation mode aerosol is an important process. Nevertheless, the discussion about the implication for the bromine recycling introduced by the neglect of the accumulation aerosol is beyond the scope of this first part of the series of articles. Here, we only discuss the simulated aerosol distribution and the pH in coarse mode aerosol. However, we will provide a detailed discussion about this issue in the third part of this series, which is dedicated to bromine release from sea salt aerosol.

Page 7220, line 21. This are 4 layers on average. This information will be added to the text.

Page 7224, Section 2.4, pages 227-228, section 2.5, and tables 1 and 2. We fully agree with the statement that global modeling imposes limits on the complexity of the scheme that can be considered. Especially deviations on a time scale shorter as the model time step (in our case 12 minutes) and smaller than the grid scale have to be parameterised or neglected. For large aerosol particles the imposed deviations fall exactly in this range. A boundary layer grid box in our model setup is approximately 250 km x 250 km wide and 65 m high. The centred height of the grid box (32.5 m) is seldomly reached by very large sea salt particles. Additionally, vertical mixing happens instantaneously by emission through the entire lowest layer. Thus the processes described in the review cannot be resolved in a global model. We therefore assumed, that on average the effect is not important, i.e. we did not include any extra parameterisation for these kind of processes. However, the existence of the large particles is not fully neglected as a lognormal distribution always accounts for some large particles.

Considering the aspect of the instantaneous mixing of fresh and aged sea salt, the effect with respect to bromine recycling can be twofold. When the amount of aged aerosol is much larger than the fresh aerosol, the amount of released bromine should be over-estimated, as the freshly emitted bromine can be released instantaneously, whereas the fresh (alkaline) aerosol needs to be acidified first. If the amount of fresh sea salt is large, the bromine release will be underestimated, as -after mixing- the aerosol might not be acidic enough to release sea salt, whereas aged aerosol itself would release bromine.

Page 7226, Sections 2.4.2 and 2.4.3 and Figure 1. Actually, both processes are treated independently in the model. Dry deposition is applied for gases and aerosols and is only calculated in the lowest model layer. Sedimentation (here used as synonym for gravitational settling) is a process important throughout the column, but for aerosols, only. Global models considering aerosols often differentiate between these processes.

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The distinction is especially important as we want to compare the results to Stier et al. (2005) who also differentiated between these two processes. Additionally, we want to show that sedimentation is the only process which does not depend on the wind. Thus we further distinguish both processes in the paper. Anyway, we deleted Sections 2.4.2 and 2.4.3 due to the comments of referee #2. We will change the caption of Figure 1.

Page 7228, lines 7-19. The discussion of the bromine aerosol chemistry itself is not the topic of this specific article. It will be discussed in detail the third part of this series.

Page 7229, line 4. Good idea, thank you.

Page 7230, lines 5-10. No, those gradients are not resolved by the model. We will add these information.

Page 7230, line 22. Yes, thank you !

Page 7230, last couple of lines As the AEROCE/SEAREX/DOE data are the only data we still compare to in the revised version and as we found no evidence that the AEROCE/SEAREX or DOE data have a cutoff diameter of 10 μm , we will delete the statement completely to avoid confusion.

Page 7231, lines 12-17. The data were obtained from AeroCom. Unfortunately, the data are not freely available, but filed in a password protected area. We are not aware of any peer-reviewed reference for the data we used, therefore we will add a personal communication to Joe Prospero.

Page 7231, lines 18-26. The sea salt mass concentrations in the data we got were estimated from the sodium concentrations by division by 0.3071. We will add this information. The stations compared in Gong et al., 1997 are already included in the comparison with the AEROCE/SEAREX/DOE data (Fig. 5). Since the analysis of the simulation is only perform for the year 2000 and since we do not have access to the longterm data, we do not want to repeat a comparison with these stations. As proposed, we will drop the EMEP comparison. and the acronyms will be explained in the acronym table.

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Page 7231, last paragraph. The data we show were provided by AeroCom as AEROCE data. Meanwhile we checked with Joe Prospero and we will be more precise about the data origins. Since we only show monthly means for the year 2000, we will add an explicit annotation that only the data for 2000 is shown.

Figure 3, legend. The lowest layer is about 52 to 70 m deep, i.e. the centre of the grid box is between 26 to 35 m high. This information will be added.

Figures 4 and 5. As proposed above, we will delete Figure 4. We will change Figure 5 and will give the information about the means accordingly.

Page 7232, line 13 to end of section. We will add an overview table, but we hesitate to shorten the discussion, as each measurement-simulation comparison shows a different aspect.

Page 7232, line 16. Yes. We will add the word cloud in the article

Page 7232, line 18. Unfortunately, M7 does not take nitrate into account, whereas the chemistry of MECCA-AERO does take it into account. We will add a comment about the neglect of nitrate in M7.

Page 7233, general. The required information will be added.

Page 7233, lines 17-22. We remove the argument about the local influences.

Page 7234, lines 26-26. Yes, you are right. These reasons are given in Keene et al., 2004. We will add this.

Page 7235, first few lines. We agree that the deviations can be positive and negative, and we will elaborate the point in more detail.

Page 7235, par. 2. It is right, that we neglected the organic acids in our simulation. We agree that organic acids may be important in some remote regions, thus we will include this argument in the discussion.

Table 4. The caption will be rephrased.

Page 7236, last paragraph, and Figure 13. We revise the text to state more precisely

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which pH is measured and which one inferred.

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